

## Evaluation of trapping parameters of thermally stimulated glow curves in KCl:Pb:Mn single crystals

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**Abstract** Amongst the various experimental techniques to study the trapping parameters associated with thermally stimulated luminescence glow curves, isothermal luminescence decay method (ILDM) is more effective for estimating order of kinetics ( $b$ ) and activation energy ( $E$ ). In this paper, the results of trapping parameters for the most prominent glow peak (peaking at  $140^\circ\text{C}$ ) of RT (Room temperature),  $\gamma$ -irradiated KCl:Pb:Mn are reported. It is established that  $140^\circ\text{C}$  glow peak of KCl:Pb:Mn is associated with first order kinetics and the activation energy is found to be  $0.54\text{eV}$ . This method is applicable not only for first order and second order kinetics but also for T S L (Thermally stimulated luminescence) processes involving intermediate order of kinetics.

**Keywords** Thermally stimulated luminescence, trapping parameters, single crystals

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In thermally stimulated luminescence studies, evaluation of trapping parameters namely order of kinetics, activation energy and frequency factor is one of the important studies in order to understand the mechanism of luminescence process on the basis of theoretical models. The works of Randall and Wilkins (1) and Garlick and Gibson (2) laid the foundations by first giving the analytical expressions for glow peaks obeying first order and second order kinetics respectively. Although there are various analytical methods (3, 4) for evaluation of trapping parameters like initial rise method, method using different heating rates, method based on glow curve shapes (Chen's method), inflexion method, area measurement method –each method has its own merits and demerits. This is because any of the analytical methods demand a completely or fairly isolated glow curves for ascertaining the order of kinetics and in turn, for determining the activation energy and frequency factor. A new (5) method namely isothermal luminescence decay method (ILDM) is more effective, reliable and versatile technique for the evaluation of trapping parameters because there are no pre-requisite conditions in this method and can be applied not only for T S L processes involving first and second order kinetics but also for

the intermediate order of kinetics. In this paper, the results of trapping parameters for the most prominent glow peak (peaking at  $140^\circ\text{C}$ ) of RT,  $\gamma$ -irradiated KCl:Pb:Mn single crystal are reported.

Laboratory grown single crystals of KCl:Pb:Mn of size ( $4\text{mm} \times 4\text{mm} \times 2\text{mm}$  approx.) were used for T S L recording and isothermal decay studies. The samples were heated at  $400^\circ\text{C}$  for 10 mins. and quenched to RT (room temperature) before exposing to gamma dose at RT from a  $^{60}\text{Co}$  source for different times as mentioned in the paper. To record T S L glow curves, the samples after exposed to  $\gamma$ -irradiation, were heated at the rate of  $150^\circ\text{K}/\text{min}$  using Indotherm-487 temperature programmer and the T S L emission was detected by an R C A 931B photomultiplier tube connected to an electrometer amplifier with the help of an Omniscribe Digilog Dual Channel recorder, the sample temperature and T S L output signal were recorded simultaneously. In the case of decay curves, the temperature was maintained at a particular temperature and the decay of T S L output with time was recorded.

The isothermal luminescence decay method (ILDM) is based on recording the decay of luminescence intensity with respect

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to time. The first step in this method, is to record the T S L glow curves and to note the glow peak temperatures. Then three temperatures are chosen around the peak temperature whose trapping parameters are to be determined. The sample is exposed to either X-ray or  $\gamma$ -rays of suitable dose. Then the sample is heated to one of the chosen temperatures and decay of the luminescence intensity is recorded at a constant temperature. This procedure is repeated for other two temperatures. The experimental data is tested for ascertaining the order of kinetics. In the case of first order and second order kinetics, the plots of  $\ln I$  vs time and  $(I_0/I)^{1/2} - 1$  vs time should yield a straight line where  $I_0$  is the intensity at  $t = 0$  and  $I$  is the intensity at any instant  $t$ . If the straight line nature is not obtained in either case, then the experimental data is fitted in the straight line nature of  $(I_0/I)^{(b-1)/b} - 1$  vs time where  $b$  is the order of kinetics ( $1 < b < 2$ ) by choosing different test values of  $b$ , among which the value of  $b$  that gives the best fit as straight line gives the order of kinetics. The slope ( $m$ ) of the straight line plots at different temperatures are noted and again  $\ln [m(T)]$  vs  $1/T$  are plotted which is a straight line (where  $T$  is the temperature in  $^{\circ}\text{K}$ ). The slope  $m' = -E/K$  of this straight line gives thermal activation energy ( $E$ ) where  $K$  is the Boltzman's constant. The theoretical details of this method are given in Ref. [5].

The T S L glow curves of KCl:Pb:Mn single crystals were recorded after  $\gamma$ -irradiating at RT for 1 min. and 10 mins. as shown in Figure 1. The glow curves exhibit three glow peaks around 90, 140 and 190 $^{\circ}\text{C}$  followed by a weak shoulder around 235 $^{\circ}\text{C}$ . Of all these glow peaks, the one at 140 $^{\circ}\text{C}$  is the strongest in intensity. Moreover, as the irradiation dose is increased, the intensities of the glow peaks are found to increase. The intensity scale for the glow curve corresponding to 10 min. irradiated sample is five times that for 1 min. irradiated sample. This shows that more and more traps responsible for these glow peaks were getting filled with the increase of  $\gamma$ -dose. Subsequently, these

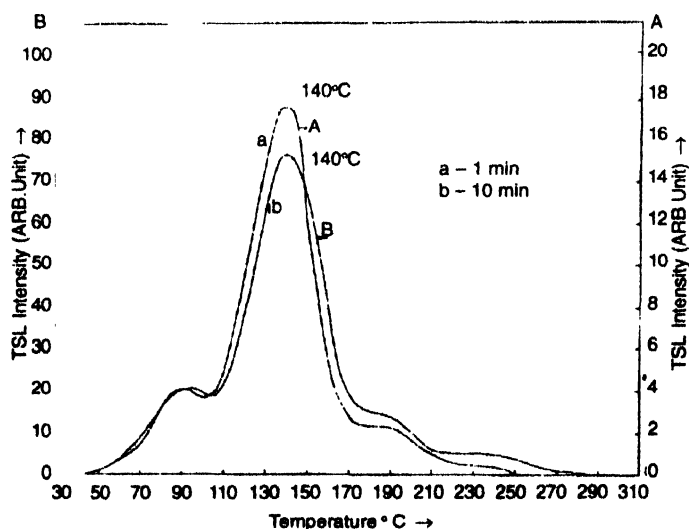


Figure 1. T S L glow curves of KCl:Pb:Mn crystal after  $\gamma$ -irradiating at RT for (a) 1 minute and (b) 10 minutes.

traps release the trapped charge carriers on the thermal stimulation which finally recombine with their counterparts giving rise to T S L glow peaks.

In order to calculate the trapping parameters corresponding to 140 $^{\circ}\text{C}$ , three temperatures namely 120, 135 and 145 $^{\circ}\text{C}$  were chosen and decay of intensity with time at these temperatures were recorded by  $\gamma$ -irradiating the crystal for 1 min. The plots of  $\ln I$  vs time were drawn at these temperatures which were straight line in nature as shown in Figure 2. This shows that the glow

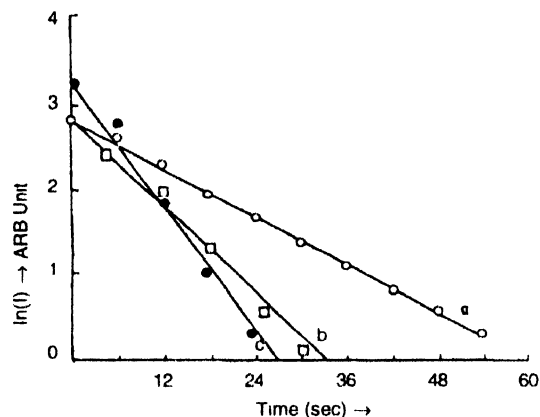


Figure 2. Isothermal luminescence decay curves of KCl:Pb:Mn crystal for 140 $^{\circ}\text{C}$  glow peak. The temperatures for (a) 120 $^{\circ}\text{C}$ , (b) 135 $^{\circ}\text{C}$  and (c) 145 $^{\circ}\text{C}$ .

peak corresponding to 140 $^{\circ}\text{C}$  obey first order kinetics. For further confirmation of the above result, the observed data was tested for intermediate order of kinetics. Plots of  $I_0/I^{(b-1)/b} - 1$  vs time were drawn for different values of  $b$  ( $1 < b < 2$ ). It was found that these plots did not exhibit straight line nature. Ultimately, the best straight line fitting was obtained for the  $\ln I$  vs time plots which established that the glow peak corresponding to 140 $^{\circ}\text{C}$  obeys first order kinetics. The slope ( $m$ ) of these straight lines for the temperatures 120, 135 and 145 $^{\circ}\text{C}$  were noted and were plotted as  $\ln(m)$  vs  $1/T$  as shown in Figure 3 which is a straight

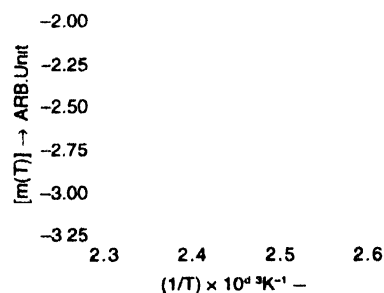


Figure 3. Plot of  $\ln [m(T)]$  vs  $1/T$  of KCl:Pb:Mn crystal for 140 $^{\circ}\text{C}$  glow peak.

line. The activation energy is calculated from the slope of this straight line ( $m' = E/K$ ) which is found to be 0.54 eV. Thus, the I L D M has revealed that the T S L glow peak at 140 $^{\circ}\text{C}$  in KCl:Pb:Mn is due to first order kinetics. This method is equally applicable for general order kinetics as reported in our earlier

papers (5, 6). Thus, the isothermal luminescence decay method is the most versatile method for the evaluation of trapping parameters among the various methods known so far.

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